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Comparison of Modelled and Measured Tracer Gas Concentrations during the Across North America Tracer Experiment (ANATEX)

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COMPARISON OF MODELLED AND MEASURED TRACER GAS CONCENTRATIONS DURING THE ACROSS NORTH AMERICA TRACER EXPERIMENT (ANATEX)

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16. ABSTRACT

The 24-hour surface concentrations of several perfluorocarbon tracer gases measured during the 1987 Across North America Tracer Experiment (ANATEX) provided an unique continental-scale data set with which to evaluate long-range transport and diffusion models. One such model, a multilayer Lagrangian model, was evaluated in the ANATEX Model Evaluation Study (AMES) by comparing distributions and time series of calculated and measured tracer concentrations at bands of sampling sites nearly equidistant from one of the two tracer release sites and by computing spatial differences in the concentration-weighted centroids of 20, 24-hour tracer "footprints" or composite plumes.

The evaluation results for this model, developed by the Air Resources Laboratory of the National Oceanic and Atmospheric Administration, indicated that it overemphasized the effects of the stronger upper-level winds: Modelled transport speeds were greater by an average of 25% and with few exceptions, modelled trajectories were to the right of those constructed from measurements. In spite of the bias in transport speed, the distributions of the calculated and measured concentrations were quite similar; with few exceptions, the values of the first three quartiles were within 20% of each other.

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COMPARISON OF MODELLED AND MEASURED TRACER GAS CONCENTRATIONS DURING THE ACROSS NORTH AMERICA TRACER EXPERIMENT (ANATEX)

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INTRODUCTION

Comparisons of the calculations of long-range transport and diffusion models with measurements on commensurate scales for extensive time periods are essential to establish levels of model uncertainty, to relate model sensitivity to model performance, and to determine the various types of meteorological scenarios for which models perform best and worst. Several recent model evaluation and sensitivity studies have used the tracer concentration data of the 1983 Cross-Appalachian Tracer Experiment (CAPTEX) (Ferber, *et al.*, 1986) to assess the performance and sensitivity of these models (Draxler, 1987; Lee, 1987; Haagenson *et al.*, 1987; Kao and Yamada, 1988; Draxler and Stunder, 1988; and Kahl and Samson, 1988).

During CAPTEX, 3-hour and 6-hour tracer concentrations were measured at the surface and tracer profiles were constructed from aircraft measurements within 1100 km of the release site up to several days after 7 tracer releases. Although this data set enabled modelers for the first time to assess the performance of long-range transport and diffusion models, the low number of tracer releases precluded definitive assessments of the models. In addition, since the CAPTEX protocol dictated that tracer only be released for homogeneous synoptic situations when the tracer would be transported across a narrow sampling sector, the CAPTEX data are basically restricted to one type of meteorological scenario.

Subsequent to CAPTEX, the Across North America Tracer Experiment (ANATEX) was conducted during the first 3 months of 1987 to provide a much more extensive data base with which to evaluate these long-range models. During ANATEX there were many more tracer releases (66); tracers were released periodically regardless of meteorological conditions; and the spatial scale of the sampling network, approximately 3000 km, was nearly 3 times greater than that of the previous study. The data base is to be used in the ANATEX Model Evaluation Study (AMES) to assess the performance of 10 long-range transport and diffusion models ranging in complexity from a single-layer Lagrangian model to a multilayer Eulerian model. This paper focuses on a discussion of the evaluation results for a multilayer Lagrangian model.

THE ACROSS NORTH AMERICA TRACER EXPERIMENT (ANATEX)

In early 1987 a team consisting of United States and Canadian scientists and technicians conducted a field study in North America to create a data base that would enable one to:

1) Determine the growth rate of plumes as they are transported beyond 1000 km of the source;

2) Describe the changes in the vertical distribution of plume mass as a function of time after release and distance from the source;

3) Investigate the effects of vertical wind shear on horizontal and vertical plume mass distributions; and

4) Verify model calculations of plume transport and dispersion, with respect to time after release and distance from the source, on a continental scale and for a wide range of meteorological scenarios.

This was accomplished by releasing distinct tracer gases from two sites in central North America (Glasgow, Montana (GGW) and St. Cloud, Minnesota (STC)) and sampling 24-hour concentrations at 77 surface sites (Figure 1) during the period from 5 January through 29 March 1987. Two inert, nondepositing perfluorocarbon tracer gases were released at the surface for 3-hour periods at 2.5-day intervals while a third was released at 5-day intervals (Tables 1 and 2); the third tracer gas served as a means to distinguish consecutive releases from STC.

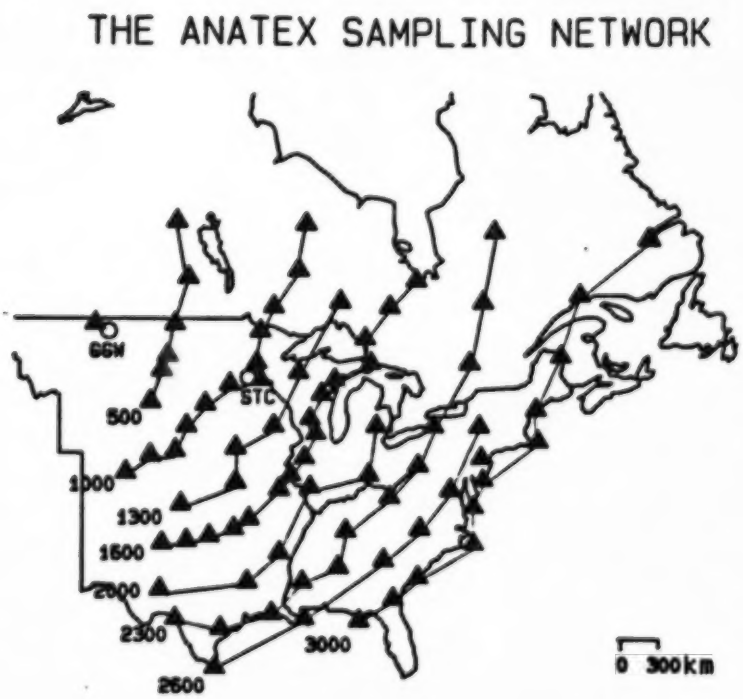

Figure 1. The location of the tracer release sites at Glasgow, Montana (GGW) and St. Cloud, Minnesota (STC) and the 77 surface sampling sites of the ANATEX network; approximate distances (km) from GGW to each band of sites are indicated.

Table 1. Attributes of Perfluorocarbon Tracer Gases Used in ANATEX.

	Perfluoromethyl-cyclohexane	Perfluoro-ortho-dimethylcyclohexane	Perfluorotrime-thylcyclohexane
Abbreviation	PMCH	PDCH	PTCH
Release Site	St. Cloud, MN	St. Cloud, MN	Glasgow, MT
Release Frequency (days)	5.0	2.5	2.5
Molecular Weight (g/mole)	350	400	450
Background (fl/l)	3.0	0.3	0.1
Purity (%)	99.0	45.0	99.0

Table 2. Perfluorocarbon Tracer Release Schedule During ANATEX.

Release Number	Date	Time (GMT)	Release	Date	Time (GMT)	Release	Date	Time (GMT)
1	Jan 5	17-20*	12	Feb 2	05-08*	23	Mar 1	17-20*
2	8	05-08*	13	4	17-20*	24	4	05-08*
3	10	17-20*	14	7	05-08*	25	6	17-20*
4	13	05-08*	15	9	17-20*	26	9	05-08*
5	15	17-20*	16	12	05-08*	27	11	17-20*
6	18	05-08*	17	14	17-20*	28	14	05-08*
7	20	17-20*	18	17	05-08*	29	16	17-20*
8	23	05-08*	19	19	17-20*	30	19	05-08*
9	25	17-20*	20	22	05-08*	31	21	17-20*
10	28	05-08*	21	24	17-20*	32	24	05-08*
11	30	17-20*	22	27	05-08	33	26	17-20*

* Simultaneous release of PMCH at STC

After laboratory analysis of the air samples, an uncertainty for each tracer concentration was calculated via the expression

$$[(\sigma_b^2 + \sigma_r^2)]^{1/2}$$

where the first term represents the variance of the gas chromatograph (GC) "background" concentrations (where these concentrations were defined as those below the 40th percentile value of each 21-day series of samples at a site); the second term represents the variance of the analyses of reference tracer gas standards from each of the three analytical laboratories.

In addition to these measurements, 12-minute samples of tracer gas were obtained from aircraft within 500 km of the release sites to define initial transport and dispersion aloft. Concentrations were also measured at 5 tower sites located approximately 1600 km from Glasgow for 6-hour periods at the surface and 6- and 24-hour periods at 200 m.

THE ANATEX MODEL EVALUATION STUDY (AMES)

AMES focuses on a performance assessment of 10 long-range transport and diffusion models across the continental scale for the entire ANATEX period as well as for specific meteorological scenarios. Models that are evaluated range in complexity from a single-layer Lagrangian model to a multilayer Eulerian model. The study also identifies those meteorological scenarios for which each model tends to perform best and worst. The information provided by this study is useful to modelers and model users to define model confidence limits, to describe model behavior for specific meteorological scenarios, and to assist in improving the models.

In general terms, the 10 models have been developed to simulate the fate of emissions along a path from the source to the ultimate receptor within a prescribed domain. Specifically, some of these models were developed to simulate surface concentrations of hazardous materials, while others were developed to simulate wet and dry deposition of acidic species. Regardless of their purpose, each model must first simulate the primary processes, namely mass transport and dispersion. With the ANATEX data base, one can evaluate these models for these primary processes.

The remainder of this paper briefly describes the model evaluated and discusses the model evaluation protocol and the results of the evaluation. The evaluation described here is restricted to the PTCH tracer released from GGW; comprehensive discussions of AMES and the ANATEX data base will be published subsequently.

Description of the Model Evaluated

One of the models evaluated in AMES is the Air Resources Laboratory (ARL) model, a multilayer Lagrangian model developed by Draxler (1987). This model uses polar stereographic grids of wind velocities derived from 6- and 12-hour rawinsonde data and interpolated to 3-hour periods. Trajectories of puffs released from GGW and STC at times and masses corresponding to the actual ANATEX tracer releases are initially calculated at a constant level above the terrain. Vertical mixing coefficient profiles, computed via an iterative solution to the normalized surface layer flux relations (Draxler, 1987) are used to determine the vertical extent of the mass. When mass reaches other vertical levels, independent puffs are created and advected using wind velocities appropriate to those levels. Horizontal dispersion is assumed to follow the linear growth suggested by Heffter (1965),

$$\sigma_h = 0.5\ t$$

while vertical dispersion is assumed to follow the relationship

$$\sigma_z = (2K_z t)^{0.5}$$

where the units are meters for σ_h and σ_z and seconds for t. Air concentrations are computed at a user-specified set of sites at a prescribed level.

Model Evaluation Protocol

Because of the 24-hour sampling periods specified by the ANATEX sampling protocol, the spatial dimensions of the tracer clouds constructed from ANATEX data were actually a composite of a consecutive series of tracer plumes and therefore, were greater than the instantaneous dimensions of the actual plume. These 24-hour composite plumes, referred to as "footprints", can have dimensions in excess of 1000 km and represent the finest level of detail of the ANATEX data base. The model evaluation protocol was designed with this in mind.

As opposed to most model evaluation studies, the AMES protocol does not compare model calculations and measurements at sites paired in both time and space. The reason for this relates to the density of the ANATEX sampling network and is explained by the following example. Suppose that there is a band of sampling sites spaced hundreds of kilometers apart and each approximately 1000 km downwind of the tracer release point. Further suppose that the model-calculated footprint is displaced by a small distance from the measured footprint so that each footprint encompasses only one of two adjacent sampling sites. A model evaluation based on a comparison of concentrations paired in both space and time would indicate that the model failed to replicate the measured data and would not distinguish this case from a case where the two footprints were very far apart. A more practical evaluation would indicate that the model performed rather well, missing the location of the measured footprint by a very small distance relative to the transport distance, and would distinguish this case from those where the footprints are far apart. The latter approach was preferred for AMES.

Rather than comparing calculations and measurements paired in space, the AMES evaluation compares time series and uses the performance measures listed in Table 3. The time series of daily mean concentrations, compared the timing as well as the magnitude of the calculated and measured concentrations at sampling sites along the bands positioned approximately 1000, 1300, 1600 and 2000 km from GGW (see Figure 1). These plots would reveal any tendency of the model to underpredict or overpredict tracer concentrations as a result of differences in modelled transport speeds and dispersion rates. When interpreting the time series plots, one must realize that when a peak calculated concentration lags a peak measured concentration by one day on the plot, this does not necessarily mean that the lag was actually 24 hours; the 24-hour sampling resolution precluded a more exact determination of the lag time.

Table 3. AMES descriptive tools and performance measures for model evaluation.

	Tools or Measures	Processes Evaluated	Concentrations Paired in Time?
1)	Time series plots of daily mean concentrations along bands of sites nearly equidistant from GGW	Transport Dispersion	Yes
2)	Percent difference between the first, second and third quartiles of the calculated and measured concentrations along bands of sites nearly equidistant from GGW	Transport Dispersion	No
3)	Difference in the locations of footprint centroids	Transport	Yes
	a) Distance separating centroids normalized by transport distance of measured footprint		
	b) Ratio of distances from source to centroids		

For the model performance measures, a PTCH threshold value of 13 dfL/L (decifemtoliters per liter or 10^{-16} L/L) was used to disregard those measured concentrations that may have relatively high uncertainties; statistical analyses of the concentrations and their uncertainties indicated that many concentrations below this threshold value were not significantly different from 0 at the 95% confidence level. For consistency, calculations below the threshold were not considered as well.

The first set of model performance measures -- the percentage difference in the values of the first, second, and third quartiles -- would reveal any tendency for the model to underpredict or overpredict concentrations along the same 4 bands of sites. Only those calculations at sites where measurements were simultaneously available were considered in the distributions.

To evaluate models strictly for the transport process, the final two performance measures assessed the differences in the location of concentration-weighted centroids of the site calculations and measurements associated with footprint pairs that could be attributed to single tracer releases. These two performance measures are based on the distances illustrated in Figure 2. The first measure ($D_{c,m}/D_{m,s}$) assessed differences in both transport direction and speed; its values were defined by the distance separating the centroids of the calculations and measurements ($D_{c,m}$) as a function of time after release and is normalized by the accumulated transport distance from the tracer source ($D_{m,s}$). Normalizing this quantity places less significance on small differences far from the release point. Values of this measure for a perfect model would be 0.

The second measure ($D_{c,s}/D_{m,s}$) assessed the differences in transport speeds of the measured and calculated footprints. The values of this measure were determined by the ratio of the distance between consecutive centroids of the calculations over that of the measurements. For a perfect model the values of this measure would be 1. Values greater than 1 would signify that the model tended to transport the footprints faster than those measured; values less than 1 would signify that the model tended to transport the footprints slower than those measured.

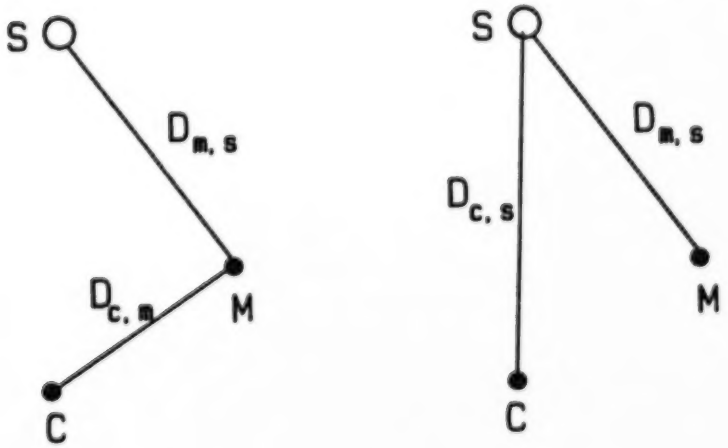

Figure 2. Definition of the distances employed in the performance measures assessing modelled transport speed and direction, where "S" is the source of the tracer, "M" and "C" are the centroids of the measured and calculated tracer footprints, and "D" is the distance between points designated by the subscripts.

Comparison of Calculated and Measured Concentrations

The comparison of the distributions of the ensemble of calculated and measured concentrations along 4 consecutive bands of sampling sites from 1000 to 2000 km of GGW (Table 4) revealed two main traits of the model performance. First, within 2000 km of GGW the calculated concentrations exceeded the threshold at 24 to 58% fewer sites than the measurements indicated; the discrepancy was not a function of distance from GGW, but appeared to be related to the number of sites along each band. Secondly, for those concentrations exceeding the threshold, the distribution of the calculated concentrations were comparable to those measured. With few exceptions, quartile values were within 20% of each other. At the 1300-km band, the differences were greater, however, because of the significant discrepancy of the sample sizes (i.e., 27 versus 65 for the calculations and measurements, respectively), the comparison of frequency distributions for this band could be misleading. These two traits indicated that the model could be overestimating vertical mixing while underestimating horizontal diffusion and/or overestimating transport speed.

The set of time series plots of daily mean concentrations for the 1000- and 1300-km bands (Figure 3) indicated that, with few exceptions, the occurrence of peak mean calculations corresponded within a day of the peaks in the mean measurements. Beyond 1300 km the correspondence of the peaks diminished. These time series plots, which consider all values regardless of whether they are in or out of the footprint, showed that the model tended to underpredict the daily mean concentrations along the 1000-, 1300-, and 1600-km bands. Although at first glance this would appear to contradict what the quartile comparisons indicated, the results would be consistent if the model footprints were indeed narrower than those measured (i.e., additional zero concentrations would reduce the mean concentration).

The comparison of the locations of the centroids for the 20 footprint pairs (Table 5) revealed several traits of the model performance in simulating tracer transport. First, as illustrated by Figure 4, the model tended to overpredict the 24-hour transport distance (i.e., $D_{c,s}/D_{m,s} > 1$). Considering only those 16 footprint pairs where the centroids were within 500 km of each other (denoted by the squares in Figure 4), the model tended to overestimate the transport distance by an average of 25%. Approximately 60% of the time, however, the calculated transport distance was within 25% of that of the measured footprints.

Table 4. Comparison of the distributions of measured (M) and calculated (C) tracer concentrations exceeding 13 dfL/L along bands of sites nearly equidistant from the release site GGW.

Approximate Distance of Band from GGW (km)	Number of Sampling Sites in Band	Sample Size and Percent Difference (D)			Quartile Values (dfL/L) and Percent Difference (D)								
					25%			50%			75%		
		M	C	D	M	C	D	M	C	D	M	C	D
1000	12	154	117	24	20	19	5	33	31	6	56	52	7
1300	6	65	27	58	15	19	27	20	27	35	37	42	14
1600	16	86	58	33	16	17	6	20	24	20	34	33	3
2000	6	51	23	55	16	16	0	20	20	0	30	32	7

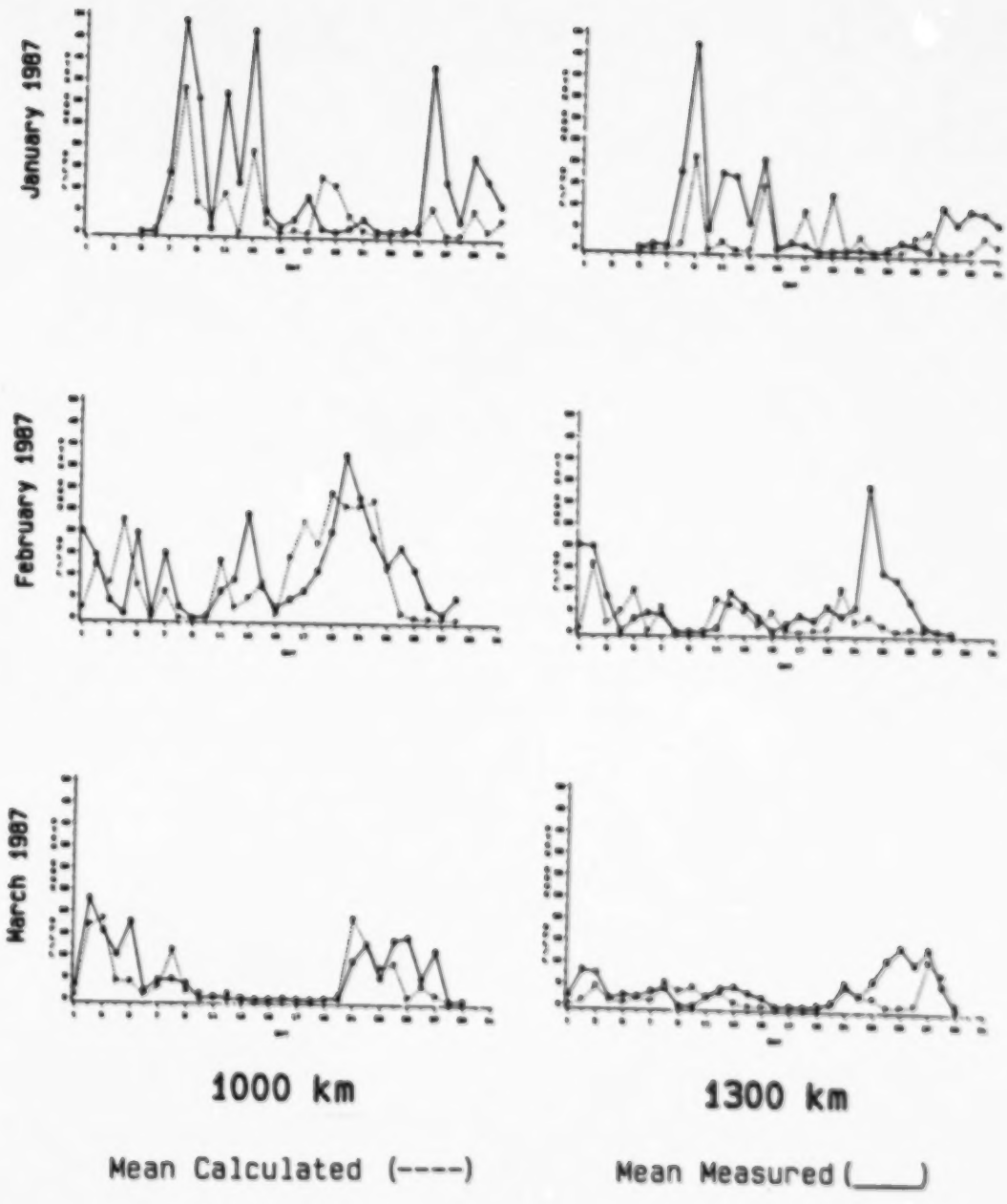

Figure 3. Time series plots comparing 24-hour PTCH concentrations averaged each day across bands of sampling sites located approximately 1000, 1300, 1600, and 2000 km from the GGW release site.

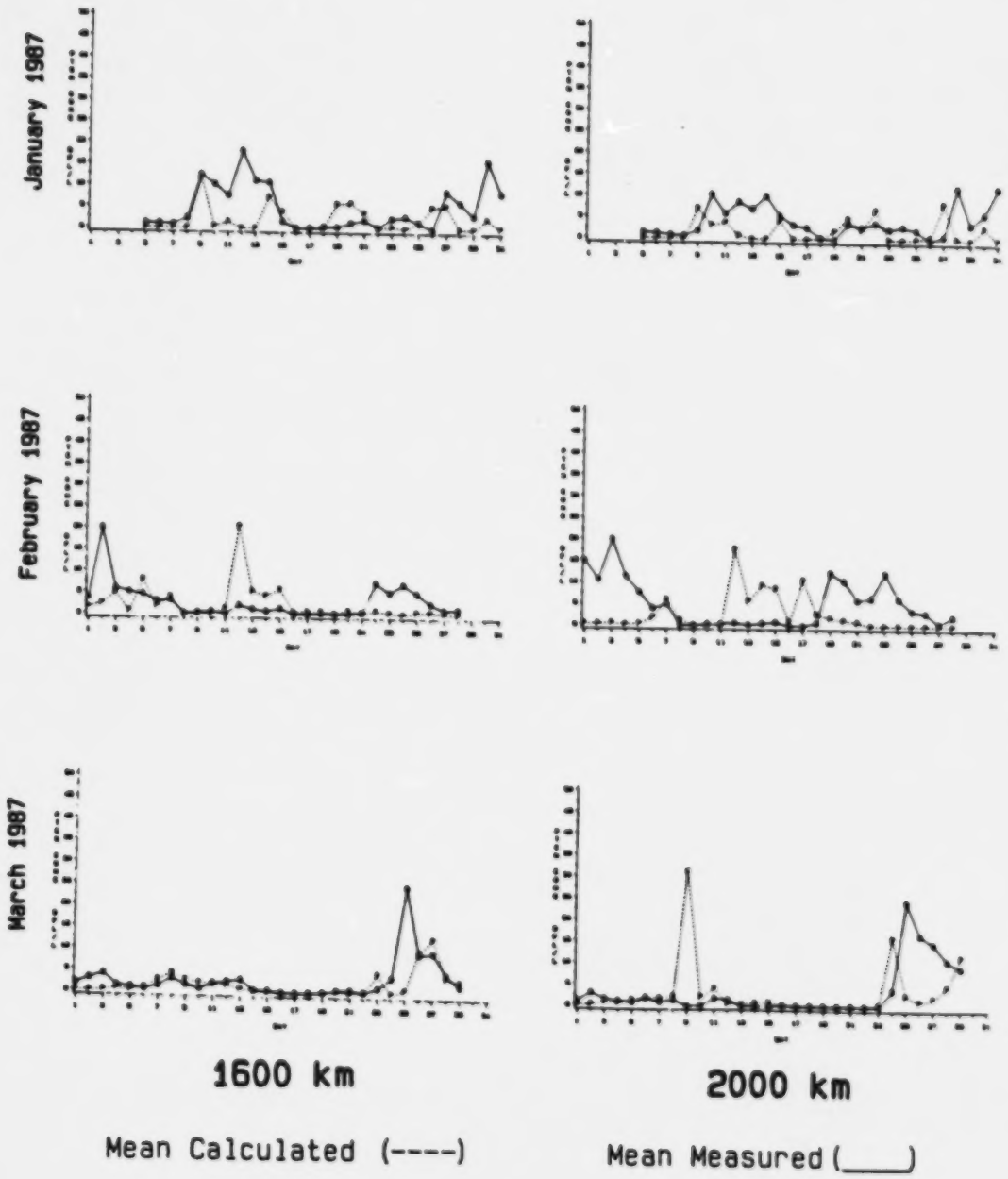

Figure 3. (Continued) Time series plots comparing 24-hour PTCH concentrations averaged each day across bands of sampling sites located approximately 1000, 1300, 1600, and 2000 km from the GGW release site.

Table 5. Distinguishable footprints used to compare calculated and measured transport speeds.

Release Number	Days After Release	24-Hour Transport Distance (km) from GGW	Geographical Location of Centroids
1	1	550	South Dakota
1	2	720	South Dakota
1	3	1150	South Dakota
1	4	1400	Iowa-Kansas
1	5	2250	Kentucky-Alabama
3	1	1040	Minnesota
4	0	630	South Dakota
4	1	1440	Kansas
4	2	2230	Texas
7	0	430	North Dakota
7	1	1020	Minnesota-Wisconsin
10	1	560	South Dakota
10	2	1140	Iowa-Illinois
10	3	1830	Missouri-Georgia
10	4	2250	Indiana-Georgia
11	1	610	Manitoba-North Dakota
11	2	1170	Ontario-Minnesota
13	1	980	Ontario-Minnesota
13	2	1570	Ontario
15	2	470	North Dakota-Minnesota

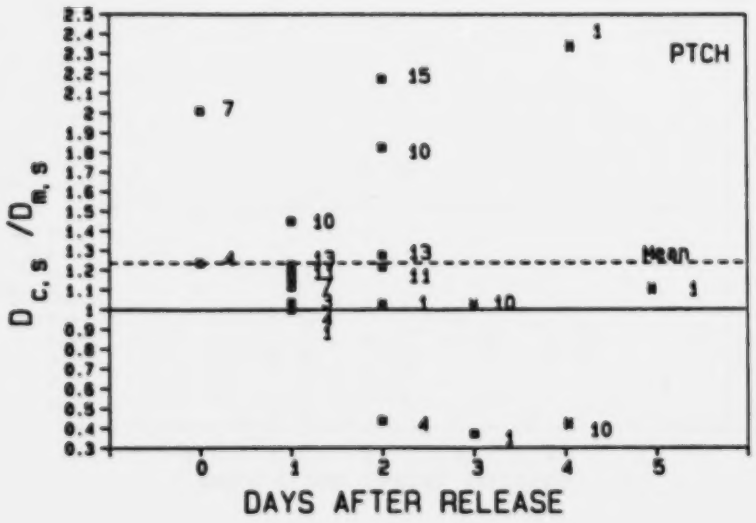

Figure 4. Comparison of calculated and measured tracer transport distances for 20, 24-hour footprint pairs where the performance measure is defined as the ratio of the distances between consecutive centroids of the calculated and measured footprints (c and m, respectively); indicated is the PTCH tracer release number and identified are those cases where the centroids of the calculated and measured footprints were greater than 500 km apart (*).

For 5 of these 16 cases, the calculated transport speeds differed substantially (i.e., >60%) from those indicated by the measurements; these differences were not attributed to large separation distances between the footprint pairs (as for those cases denoted by the asterisks in Figure 4). On the initial day of release 7 and the second day of release 10, the model transported the tracer approximately twice as far as the measurements indicated; evidently the calculated wind speeds were influenced more by the stronger winds aloft while the measurements indicated that the actual transport speed was restricted to the much lighter wind speeds near the surface. This could be a result of the model exaggerating the vertical mixing of the tracer. Conversely, calculated wind speeds for the second day of release 4 and the third day of release 1 underestimated the wind speeds by approximately 40%; for these cases the tracer was located near a weak cold front and within a ridge of high pressure, respectively.

Another trait revealed by the comparison was that the relative errors in simulating the footprint centroids ($D_{c,m}/D_{m,s}$) generally ranged from 20 to 60% of the transport distance of the measured footprint (Figure 5). Relative errors exceeded the transport distance of the measured footprints for 2 cases, the initial day of release 7 and the second day of release 15. These large errors were attributed to overestimates in the transport speed. Finally, with few exceptions, the trajectories of the calculated footprints were to the right of the trajectories of the measured footprints, an indication that the model is overemphasizing the significance of the upper-level winds.

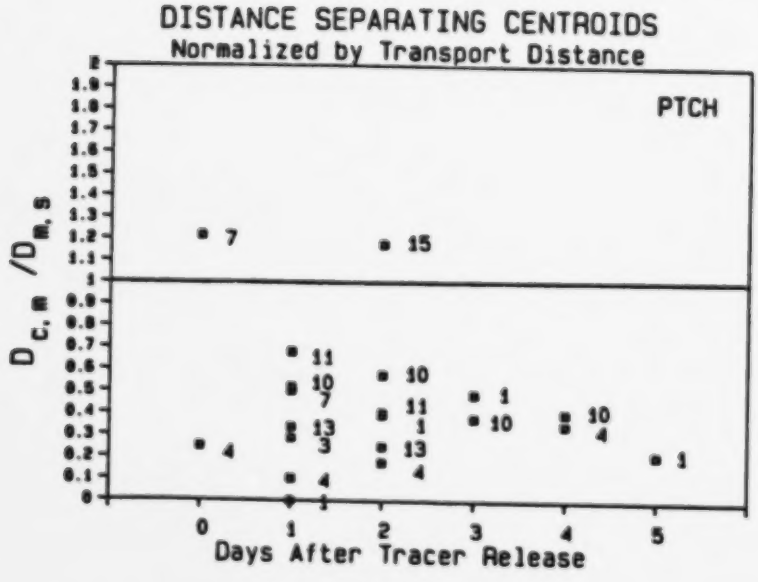

Figure 5. Comparison of calculated and measured transport direction and speed for 20 footprints where the model performance measure is defined by the ratio of the distance separating the centroids of the calculated and measured footprints over the total transport distance of the measured footprint centroid; indicated is the number of the PTCH tracer release from GGW.

SUMMARY

The 24-hour surface tracer concentrations of several perfluorocarbon tracer gases measured during the Across North America Tracer Experiment (ANATEX) provided an unique continental-scale data set with which to evaluate long-range transport and dispersion models. One such model, a multilayer Lagrangian model, was evaluated in the ANATEX Model Evaluation Study (AMES) by comparing distributions and time series of calculated and measured tracer concentrations at bands of sampling sites nearly equidistant from the tracer release site and by computing spatial differences in the concentration-weighted centroids of 20, 24-hour tracer "footprints" or composite plumes.

The evaluation results for the ARL model indicated that it tended to overestimate transport distances by an average of 25%. However, 60% of the time, the calculated transport distance was within +25% of the measured transport distance. The slight overestimate of the transport distance could be attributed to the model's overemphasis of the significance of upper-level wind speeds in determining the tracer transport. This could be the result of the model exaggerating the vertical mixing of the tracer. The distribution of calculated concentrations were comparable to those measured; with few exceptions, the values of the first, second and third quartiles were within 20% of each other. With few exceptions, the distance separating centroids of calculated and measured footprints was 20 to 60% of the transport distance of the measured footprints and the trajectories of the calculated footprints were to the right of those of the measured footprints. Errors in the simulated timing of frontal passages and the model's overemphasis of the significance of upper-level winds appeared to explain those cases when centroid pairs were farthest apart.

Subsequent publications will include comprehensive discussions of the ANATEX data base and the evaluation results of this and the other 9 models participating in the ANATEX Model Evaluation Study.

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